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16. Abstract

In multi-component ionic diffusion systems, uphill diffusion is frequently observed. The conditions for the development of uphill diffusion were investigated by computer simulation of ion exchange between a glass containing two cations (A, B) and a molten salt containing the third cation (C). The following conclusions are obtained. (1) The uphill diffusion of B ion is observed when $D_A > D_B > D_C$ or $D_A > D_C > D_B$, if all diffusion coefficients D's are independent of concentration. (2) This uphill diffusion behavior is affected by the molar ratio of A to B in a mother glass. In the simulated ion exchange systems that were investigated, the uphill diffusion was prominent when the molar fraction of B ion is around 0.25. (3) The concentration dependence of D does influence the distribution of ions, however, the relationship between three diffusion coefficients is more important condition for the development of the uphill diffusion. Ion exchange experiments in (Na, Rb) glass/(K) salt and (Na, K) glass/(Tl) salt systems were carried out to confirm the results of simulation. The uphill diffusion of Rb ion and K ion was observed as predicted by the simulation. The diffusion behavior of ions in glass during the ion exchange process for the fabrication of gradient index lens was also investigated by the computer simulation. The progression of ion concentration profiles during this ion exchange process, especially the development of the uphill diffusion of Tl ion in the initial stage of this process, was well reproduced in this simulation study.

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COMPUTER SIMULATION OF UPHILL DIFFUSION IN TERNARY IONIC SYSTEM

* /1206

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In multi-component ionic diffusion systems, uphill diffusion is frequently observed. The conditions for the development of uphill diffusion were investigated by computer simulation of ion exchange between a glass containing two cations (A, B) and a molten salt containing the third cation (C). The following conclusions are obtained. (1) The uphill diffusion of B ion is observed when $D_A > D_B > D_C$ or $D_A > D_C > D_B$, if all diffusion coefficients D's are independent of concentration. (2) This uphill diffusion behavior is affected by the molar ratio of A to B in a mother glass. In the simulated ion exchange systems that were investigated, the uphill diffusion was prominent when the molar fraction of B ion is around 0.25. (3) The concentration dependence of D does influence the distribution of ions, however, the relationship between three diffusion coefficients is more important condition for the development of the uphill diffusion. Ion exchange experiments in (Na, Rb) glass/(K) salt and (Na, K) glass/(Tl) salt systems were carried out to confirm the results of simulation. The uphill diffusion of Rb ion and K ion was observed as predicted by the simulation. The diffusion behavior of ions in glass during the ion exchange process for the fabrication of gradient index lens was also investigated by the computer simulation. The progression of ion concentration profiles during this ion exchange process, especially the development of the uphill diffusion of Tl ion in the initial stage of this process, was well reproduced in this simulation study. [*Received March 15, 1986*]

Key words : *Computer simulation, Ionic diffusion, Uphill, Ion exchange, Gradient index lens*

1. Introduction

An example of ion exchange or ion diffusion phenomenon of glass applied in industrial use is fabrication of gradient-index lenses. In this method, the glass containing monovalent cations such as thallium, cesium and etc. which increase the index of refraction of the glass is soaked in a molten salt such as potassium nitrate and for instance, the parabolic distribution of thallium ion is formed in the glass by ion exchange of thallium ions in the glass with potassium (Reference 1). There is a report concerning diffusion behavior in the fabrication process of gradient-index lenses by Kitano and his staff (Reference 2) but in the report, the relationships between the behavior and the ion concentration distribution in the glass is not studied. Thus, the authors studied the transition of the concentration distribution of monovalent cations in time lapse during the fabrication process of the gradient-index lenses containing thallium ion. In the study, the authors observed the unique concentration distribution which seems to be the uphill diffusion of thallium ion.

There are some reports (Reference 3 to 5) for the phenomenon in which the specific diffusion species diffuses in the opposite direction from the predicted direction of the initial concentration gradient; i.e. uphill diffusion, in the multi-component ionic diffusion systems having a difference in the diffusion speed of the diffusion species.

* Numbers in margin indicate pagination of foreign text.

It is presumed that the previously mentioned diffusion behavior of thallium ion is similar behavior to the above. There are a few reports (Reference 6) concerning the uphill phenomenon in ionic diffusion in the temperature range in which the frame of glass seems to be fixed substantially as in the case of the ion exchange of the solid glass with molten salt. The authors developed a simple diffusion model which can adopt the composition dependency of the diffusion coefficients for the ion exchange systems and studied the diffusion behavior of ions during the ion exchange process of glass using computer simulation.

The conditions for the uphill diffusion of ions were studied by computer simulation in the case that the glass containing two kinds of monovalent cations is soaked in the molten salt of the third monovalent cation for the ion exchange treatment and it was compared with the results of the diffusion experiment. Also, the results of simulation by time lapse of the ion concentration distribution observed in the process of the real gradient-index lens containing thallium ion are reported.

2. Diffusion model

2.1 Ion diffusion model

The diffusion of monovalent cations in glass is the diffusion of electrified particles and it is considered that the local field caused by the difference of the mobility of the diffusion species occurs and the diffusion is influenced. When it is assumed that the diffusion occurs only in x direction in the thermodynamically ideal system, the flux density of i ion is written as follows (Equation (1)) /1207
(Reference 7 and 8) from Nernst-Planck's equation.

$$J_i = -D_i \frac{\partial C_i}{\partial x} - D_i \cdot C_i \frac{Z_i \cdot F}{RT} \cdot \frac{\partial \phi}{\partial x} \qquad (1)$$

where J_i, D_i, C_i and Z_i are the flux density, the self-diffusion coefficient, the concentration and the electric charge number of i ion respectively and F is the Faraday constant, R is the gas constant, T is the absolute temperature and ϕ is the internal electrostatic potential.

Three kinds of monovalent cation A, B and C participate in the diffusion and it is assumed that the stoichiometric relation and the relation of electrical neutrality are maintained in the system.

$$Z_a = Z_b = Z_c = Z = 1 \qquad (2)$$

$$J_a + J_b + J_c = 0 \qquad (3)$$

$$C_a + C_b + C_c = 1 \qquad (4)$$

Here, the flux density and concentration of ions are handled as the normalized value for all of the monovalent cation concentration in the system.

Equation (1) is applied for each A, B and C and added together and equation (5) is obtained by applying equation (3)

$$\frac{ZF}{RT}\frac{\partial \phi}{\partial x}=-\frac{D_A\cdot\frac{\partial C_A}{\partial x}+D_B\cdot\frac{\partial C_B}{\partial x}+D_C\cdot\frac{\partial C_C}{\partial x}}{D_A\cdot C_A+D_B\cdot C_B+D_C\cdot C_C} \qquad (5)$$

When equation (5) is substituted with equation (1) for A, equation (6) is obtained.

$$J_A=-D_A\cdot\frac{\partial C_A}{\partial x}+D_A\cdot C_A \cdot\frac{D_A\cdot\frac{\partial C_A}{\partial x}+D_B\cdot\frac{\partial C_B}{\partial x}+D_C\cdot\frac{\partial C_C}{\partial x}}{D_A\cdot C_A+D_B\cdot C_B+D_C\cdot C_C} \qquad (6)$$

When the concentration, concentration gradient and diffusion coefficient for each A, B and C are given, the flux density of A ion at a point in the system can be calculated from equation (6).

2.2 Simulation operations

In this paragraph, the model of the case of the ion exchange treatment by soaking sheet glass with thickness 2L into a molten salt is described.

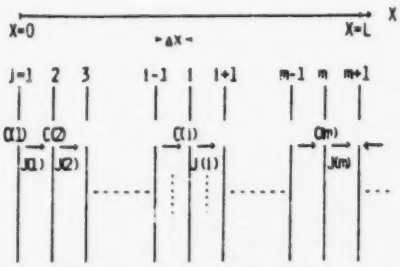

Fig. 1. Diffusion model layout.

As shown in Fig.1, a sheet with 2L of thickness and infinite length in y and z direction is considered and it is divided into thin layers numbering 2m (thickness $\Delta x=L/m$). The concentration is defined at the thin layer boundary surface and the flux density is defined at the center of the thin layer. It is assumed that the diffusion coefficient is given as the following equation with the function of the concentration C_A, C_B and C_C.

$$D_k=f(C_A, C_B, C_C),\ k=A,B,C \qquad (7)$$

At first the flux density is calculated from the concentration $C_k(j,t)$ at each boundary surface at time t with equation (6).

In the calculation of equation (6), $\partial C_k / \partial x$ is approximated with the difference of $(C_k(i+1,t)-C_k(i,t))$. For C_k of equation (6), the arithmetical mean value of $C_k(i+1,t)$ and $C_k(i,t)$ is used. Also by using this C_k, D_k is calculated with equation (7) and substituted into equation (6). From the above operations, the flux density $J_k(i)$ at the plane which is the middle of i plane and (i+1) plane is calculated.

Next, the concentration change of k ions, in the thin layer of ΔX the center of which is i plane is obtained from $J_k(i)$ and $J_k(i-1)$ calculated in a similar manner to the calculation of $J_k(i)$. Since the increase of k ions in the thin layer in the small time Δt is expressed as $(J_k(i-1)\ \Delta t - J_k(i)\ \Delta t)$, the increase $\Delta C_k(i,t)$ of the concentration of k ions can be calculated with equation (8).

$$\Delta C_k(i,t)=\frac{(J_k(i-1)-J_k(i))\cdot\Delta t}{\Delta X} \qquad (8)$$

The concentration of k ions, $C_k(i, t+\Delta t)$, at i plane at time $(t+\Delta t)$ is calculated with equation (9)

$$C_k(i,t+\Delta t)=C_k(i,t)+\Delta C_k(i,t) \qquad (9)$$

By repeating this operation, the concentration from $C_k(2)$ to $C_k(m)$ can be calculated. A constant value is obtained from $C_k(1)$ as the boundary condition and the concentration $C_k(m+1)$ at the middle of the sheet can be calculated from equation (10) which is obtained from the symmetric condition of the system.

$$C_k(m+1,t+\Delta t)=C_k(m+1,t)+\frac{2\times J_k(m)\cdot\Delta t}{\Delta X} \qquad (10)$$

By repeating the calculation of the concentration in the time lapse Δt from the given initial concentration $C_k(j, t=0)$, the concentration distribution of the ions at a certain time can be calculated.

In the case of the cylindrical shape, it is necessary to consider the concentric cylindrical thin layer; i.e. it is necessary to consider for the calculation of the ion amount change and the concentration change in the thin layer that the cross section area of the thin layer is the function of its location. Except this, the above mentioned operations can be applied for the calculation. Here, the 3 component system was discussed but the system with more than 4 components can be handled by following the simulation operations.

3. Results of diffusion simulation

3.1 The case of constant diffusion coefficient

The authors studied the case of the assumption that the diffusion coefficient of each ion is not dependent upon its composition.

The case of the ion exchange between the mother glass containing ion A and B in equimolars and a molten salt containing only ion C was assumed.

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In this case, the initial condition and the boundary condition are given by equation (11).

$$\left.\begin{array}{llll} t=0 & C_A=C_B=0.5 & C_C=0 & (x>0) \\ t>0 & C_A=C_B=0 & C_C=1 & (x=0) \end{array}\right\} \quad (11)$$

The calculation was executed at Δt = 10s with L = 0.075cm, m = 150.

Fig. 2 shows the calculation results of the case that the difference between D_A and D_B ($D_A > D_B$) is changed with constant of $D_C = 10^{-9}$ cm²/s. The case of (1), $D_A = D_B = D_C$, is the case that the analytical solution forms the error function (Reference 9) and the result of (1) in Fig. 2 coincides to the analytical solution. In the case that there is a difference between D_A and D_B, the phenomenon occurs that the concentration is higher than the original concentration in the distribution of ion B of which diffusion coefficient in the glass is smaller in the original glass. This is uphill diffusion in which the diffusion occurs in the opposite direction to the predicted direction from the concentration gradient. When the difference between D_A and D_B increases, the uphill increases as can be seen in Fig. 2.

Fig. 3 shows the concentration distribution change of B ion in the case of various D_C keeping $D_A=10^{-8}$ cm²/s, $D_B=10^{-10}$cm²/s. The prominence in the B ion distribution increases when D_C decreased until D_C reaches a certain value, then the prominence decreases.

Fig.4 shows the results of the above state and when D_C is about the same or slightly smaller than the diffusion coefficient of ion B which is the slower ion in the glass; uphill diffusion of ion B occurs remarkably.

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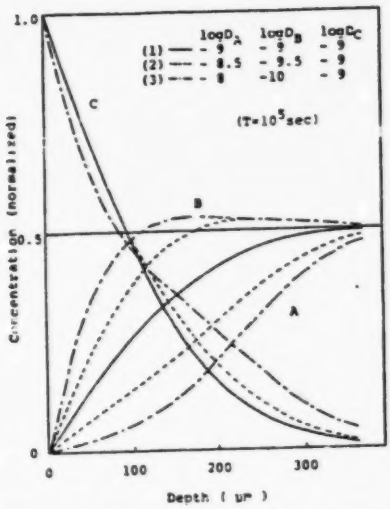

Fig. 2. Distributions of ions for various D.

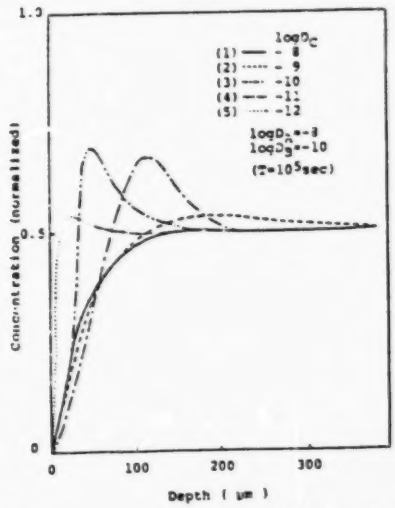

Fig. 3. Concentration profiles of B ion for various D_C.

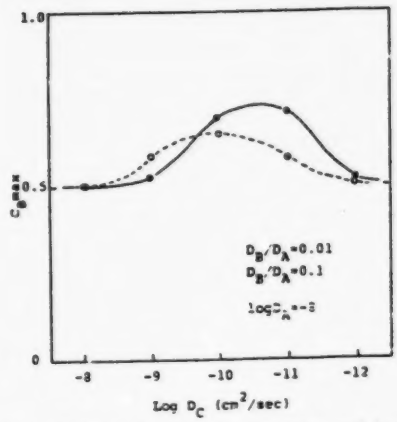

Fig. 4. Relationship between C_B max and log D_C.

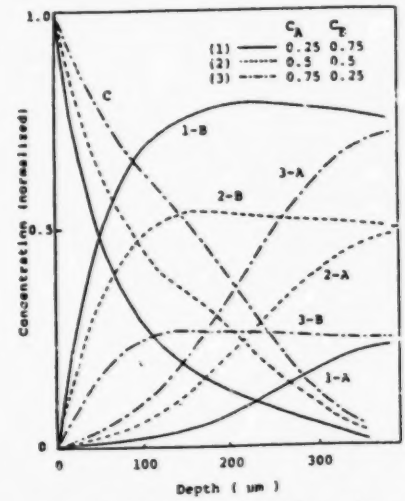

Fig. 5. Distributions of ions for various C_A/C_B($T=10^5$ s).

3.2 Effects of C_A/C_B ratio

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Effects of variation of the concentration ratio of ion A and B in a glass on the ion concentration distribution was investigated.

Fig.5 shows the results of the ion concentration distribution in the case of the various ratios of C_A and C_B, keeping $D_A=10^{-8}$, $D_B=10^{-10}$ and $D_C=10^{-9}cm^2/s$.

Fig.6 shows the relationships between the ratio of C_A and C_B and the state of the prominence on the concentration distribution of B ions. The maximum prominence occurs at around $C_B=0.25$.

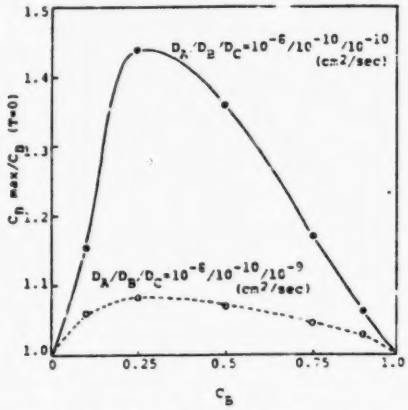

Fig. 6. Relationship between C_B max/$C_B(T=0)$ and mo ar fraction of B ion.

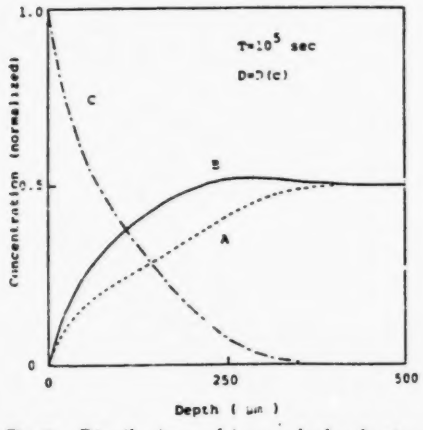

Fig. 7. Distributions of ions calculated using concentration-dependent D given by Eq. (12).

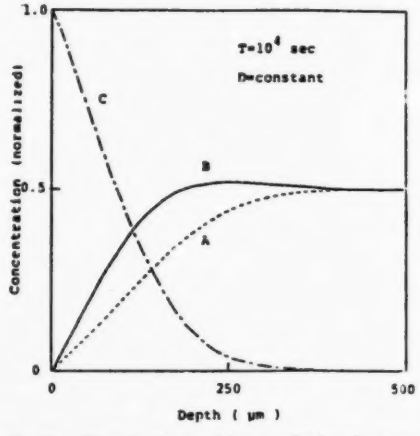

Fig. 8. Distributions of ions calculated using concentration-independent D.

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3.3 The case of diffusion coefficient dependence upon composition

It is well known that the self-diffusion coefficient of alkali-ions is much dependent upon the concentration of alkali-ions in the case that more than 2 kinds of alkali-ion exist (Reference 10 and 11). The dependency varies by composition of the glass, combination of the ions and it gets more complex when the number of the types of ion increases (Reference 12). Thus it seems that there are many discussions about the type of function for setting the dependency upon the composition. In this report, the calculation results with an example which the authors set is shown and compared with the case that the diffusion coefficient is kept constant.

Fig.7 shows the concentration distribution which was calculated with the diffusion coefficient given by equation (12).

$$\left.\begin{aligned} D_A &= 2\times10^{-8}\times10^{-1.1C_B-1.8C_C}\ \mathrm{cm^2/s} \\ D_B &= 5\times10^{-9}\times10^{-2.0C_A-1.4C_C}\ \mathrm{cm^2/s} \\ D_C &= 6\times10^{-9}\times10^{-1.8C_A-0.3C_B}\ \mathrm{cm^2/s} \end{aligned}\right\} \qquad (12)$$

Fig.8 shows the calculation results using $D_A=2\times10^{-8}$, $D_B=5\times10^{-9}$ and $D_C=6\times10^{-9}\mathrm{cm^2/s}$. The constant value of the diffusion coefficient used in the case shown in Fig.8 is the maximum value of the diffusion coefficient given by equation (12) for the ions. On the other hand, the diffusion coefficient of the ions was set so that it decreases uniformly when the ion concentration decreases in the case of Fig.7. In Fig.7 and Fig.8, the diffusion depth is about the same but the diffusion time is different and the reason that the exchange speed is faster in the case that the diffusion coefficient is constant seems to be from the above reason.

There is a difference in the process (curvature of the curve) of the distribution of A and B ions near the surface and occurrence of the effects of the composition on the diffusion coefficient is found but the basic pattern of the concentration distribution is about the same.

When the diffusion coefficients at the surface ($C_C=1$, $C_A=C_B=0$) are calculated by equation (12), $D_A=2\times10^{-9.6}$, $D_B=5\times10^{-10.4}$ and $D_C=6\times10^{-9}\ \mathrm{cm^2/s}$ ($D_C>D_A>D_B$) are obtained. On the other hand, $D_A=2\times10^{-8.55}$, $D_B=5\times10^{-10.2}$ and $D_C=6\times10^{-10.05}\mathrm{cm^2/s}$ ($D_A>D_C>D_B$) at the front portion of the diffusion ($C_C=0$, $C_A=C_B=0.5$). Thus, the relationship of the magnitude of the diffusion coefficient of each ion does not match to the condition of occurrence of the uphill diffusion of ion B mentioned in paragraph 3.1 but it seems that it matches to the condition in the low concentration region. The prominence in the concentration distribution of ion B is the phenomenon which

occurs in the high concentration region (the low concentration region of ion C) of ion B. As mentioned above, also in this diffusion system, the condition for the uphill diffusion is satisfied in the low concentration region of ion C. The similar formation of the uphill distribution of ion B shown in Fig. 7 and Fig. 8 seems to be indicating that the relationship of the magnitude of the diffusion coefficient /1210 in the low concentration region of ion C is a controlling factor of the uphill diffusion.

3.4. An example of diffusion in cylindrical shape

The calculation results of the ion distribution by using the diffusion coefficient given by equation (12) with radius, R=0.1cm, number of divisions in the radius direction, m=100 are shown in Fig. 9. In the calculation, Δt was fixed as 25s. In the case of short diffusion time, the prominence (Fig. 9(a)) in B ion distribution by the uphill diffusion is observed to the similar to the case of the ion concentration distribution of the flat plate shown in Fig. 7. When the diffusion time gets longer, the location of the prominence of B ion moves toward the center of the cylinder (Fig. 9(b)). When the diffusion time is long enough, the prominence disappears and the distribution of B ions changes into a smooth parabolic curve (Fig. 9(c)). Fig. 9(d) shows the calculation results with the constant diffusion coefficient which is the same as the case shown in Fig. 8 and it shows basically the same pattern of ion distribution as Fig. 9(a).

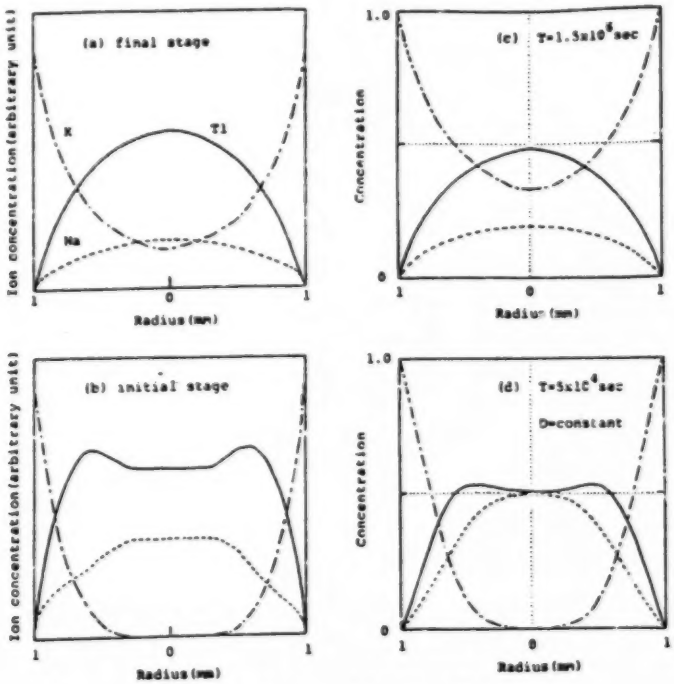

Fig. 9. Distributions of ions in cylinders.
(a), (b), (c): Concentration-dependent D
(d): Concentration-independent D

4. Comparison with experiment results of diffusion

4.1 Glass containing Rb

The measurement results by X-ray micro-analyzer (XMA) of the concentration distribution of alkali-ions after soaking a sheet glass containing $60SiO_2 \cdot 20ZnO \cdot 10Rb_2O \cdot 10Na_2O$ (mixing composition : mol%) into KNO_3 salt at 600°C in 168 hours are shown in Fig. 10.

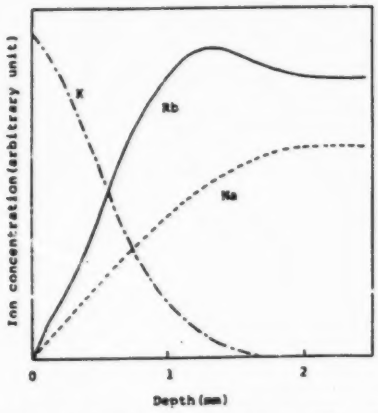

Fig. 10. Concentration profiles in (Rb, Na) glass treated with KNO_3 salt at 600°C for 168 h.

By considering the report concerning the self-diffusion coefficient of alkali-ions (Reference 10 and 11), it can be presumed that the order of the self-diffusion coefficient in the single alkali glass is Na>K>Rb in the combination of Rb, Na and K ion.

When A=Na, B=Rb and C=K. $D_A > D_B$ and D_C is between D_A and D_B in the combination of the ions in the diffusion experiment and it seems to meet the condition of occurrence of uphill diffusion of ion B. In this diffusion experiment, the prominence in /1211 the concentration distribution of Rb ion occurred as expected, as shown in Fig. 10.

4.2 Glass containing Na/K

The ion exchange treatment between the three kinds of glasses containing $60SiO_2 \cdot 5B_2O_3 \cdot 15ZnO \cdot 20R_2O$ (mixing composition : mol%) where R_2O is ($5Na_2O+15K_2O$) or ($10Na_2O+10K_2O$) or ($15Na_2O+5K_2O$) and sulfate (Tl_2O_4 : $ZnSO_4$ = 1 : 1) at 520°C for 192 hours was carried out. Fig. 11 shows the concentration distribution of each ion in the thickness direction of the glass specimen.

According to the report, (Reference 13) Tl ion is not an alkali-ion but when it coexists with alkali-ion, Tl ion shows the mixed cation effects which is similar to the mixed alkali effect and according to the report (Reference 14) of the diffusion coefficient of Tl ion, Tl ion can be considered as an alkali-ion which has a similar degree of diffusion coefficient as the Rb ion.

Now, when A=Na, B=K and C=Tl, the state of $D_A > D_B > D_C$ is presumed and it meets the conditions where uphill diffusion is expected. In the glass containing ($15Na_2O + 5K_2O$), $C_B=0.25$ and from the results of Fig. 6, observation of remarkable uphill diffusion of K ion is expected. The results shown in Fig. 11 seem to support the prediction.

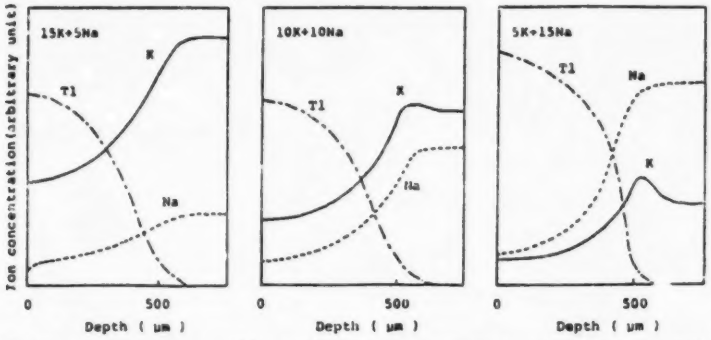

Fig. 11. Concentration profiles in (Na, K) glasses treated with ($Tl_2SO_4 + ZnSO_4$) salt at 520°C for 192 h.

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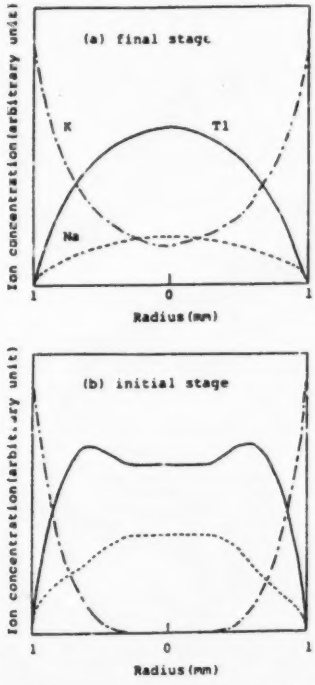

Fig. 12. Concentration profiles in (Tl, Na) glasses for gradient-index rod lens treated with KNO_3 salt.

4.3 Process for lens

Fig. 12(a) shows the concentration distribution of monovalent cation measured with XMA in the cross section radius direction of the gradient-index lens (2mm diameter) obtained from the ion exchange treatment between SiO_2-B_2O_2-Na_2O-Tl_2O glass and KNO_3 salt at 530°C for 360 hours. The distribution of the smooth parabolic curve of thallium ion in the radius direction can be seen. Fig. 12(b) shows the ion distribution of the specimen in its radius direction which had the ion exchange treatment for 72 hours. In this figure, the prominence which is equivalent to about 15% of thallium concentration in the mother glass is observed in the thallium ion distribution.

The mother glass for the lenses contains Na and Tl as the monovalent cations with about equal molar ratio. The case of the ion exchange treatment of the glass with KNO_3 salt seems to correspond to the case of the simulation of the diffusion in the cylinder, the calculation results of which were shown in Fig. 9. The diffusion coefficients (equation (12)) used in the calculations in Fig. 9(a) to (c) were estimated from the results of the counter diffusion coefficients obtained from the counter diffusion experiment of the two component system of Na-K, Na-Tl, K-Tl and they were used as Na=A, Tl=B and K=C.

The results of the diffusion simulation shown in Fig. 9 seem to well simulate /1212
the mutual relations of the concentration distribution of each ion, the state of the concentration distribution by time lapse and especially the uphill behavior of thallium ion. By comparison of Fig. 9(a) (diffusion coefficient depending on composition)and Fig. 9(d) (constant diffusion coefficient), the relative relation of the concentration distribution of the three kinds of ions is about the same but the case of Fig. 9(a) in which the dependency on composition is considered seems to better reflect the real concentration distribution of the ions (Fig. 12 (b)).

The above comparison results with some diffusion experiments indicate the possibility of comprehension of the uphill behavior qualitatively by the diffusion simulation mentioned in this report. For better quantitative investigation of the diffusion model, it is necessary to study with highly reliable self-diffusion coefficients and it will be a future study.

5. Conclusions

The authors developed a simple ion diffusion model which can count on the composition dependency of the diffusion coefficient and studied the ion diffusion

behavior during the ion exchange process in glass by computer simulation. In the case of ion exchange treatment of glass containing two kinds of monovalent cations (A, B) in a molten salt of the third monovalent cation, the conditions for the uphill diffusion of ions were studied and the follwing conclusions were reached.
(1) When the diffusion coefficient D is constant, regardless of the composition, the uphill diffusion behavior in ion B is observed in the case that the diffusion coefficient of each ion has the relations of $D_A > D_B > D_C$ or $D_A > D_C > D_B$.
(2) The degree of the uphill behavior of ion B is affected by the molar ratio of ion A and B in glass. In this study, the prominence occurs around 0.25 of the molar fraction of ion B.
(3) The composition dependency of the diffusion coefficient affects the ion concentration distribution and the occurrence of the uphill diffusion depends upon the magnitude of the diffusion coefficient of each ion.

To support the simulation results, the ion exchange experiments with the combinations of (Na, Rb) glass/(K) molten salt and (Na,K) glass/(Tl) molten salt were carried out and as expected, the uphill diffusion of the Rb ion and the K ion was observed. Also the simulation of the transition of ion concentration distribution during the fabrication process of the gradient-index lens containing Tl ion was tried and compared with the experiment results and it was verified that the diffusion simulation is useful.

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